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REACTION RATE DATA.

Number 6.
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RESUME OF EY 77

DNA-SPONSORED CHEMISTRY/PHYSICS

REACTION RATE RESEARCH PROGRAMS,

Number 60

This issue of the DASIAC Reaction Rate Data presents brief resumes of DNA-sponsored Chemistry/Physics Reaction Rate Research efforts for FY 1977.

Since information in this document is considered to be preliminary in nature and may be subject to possible future revision and/or changes, it is requested that recipients do not cite or reference the contents in other media without receipt of prior specific approval of the Defense Nuclear Agency.

Future editions of the DASIAC Reaction Rate Data will contain related progress reports concerning the DNA-sponsored efforts described herein.

Submission of other pertinent information of a related nature deemed appropriate for publication in future editions of the DASIAC Reaction Rate Data is welcome and should be sent to DASIAC, General Electric Company—TEMPO, 816 State Street, Santa Barbara, California 93102, which is contractually engaged by the Defense Nuclear Agency for this purpose.

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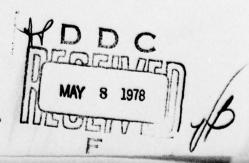
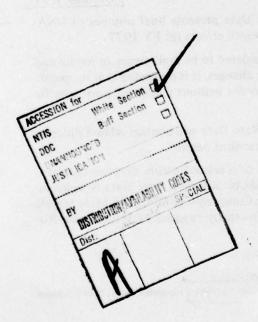




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RESUME OF FY 77 DNA-SPONSORED CHEMISTRY/PHYSICS REACTION RATE RESEARCH PROGRAMS

A. SUBTASK S99QAX HD 411

"Reaction Rates Essential to Propagation."

1. Applications of the ARICHEM Computer Code-F. Niles and J. Heimerl, AASLNM and BRL. Ends 30 September 1977 (Work Unit 01).

The following tasks will be completed in FY77:

- Predictions of "Effective Rate Coefficients" will be obtained via the AASLNM/BRL developed "AIRCHEM" computer code. Comparisons of calculated, selective ion number density parameters will be compared with laboratory experimental data in order to improve and restructure atmospheric deionization in lower and middle D region models. Relevant data from field measurements acquired during naturally occurring energetic events (such as solar proton events (PCAs); magnetospheric substorms; eclipses; auroras; etc.) will also be utilized during such comparisons.
- Calculations will be made of changes resulting in electron density variations which also
 occur in the lower D region at critical times and comparisons with actual instrumental data will be given (e.g., the solar proton event data of 1972 will be employed, as will
 other events).
- In addition, AASLNM and BRL capabilities will continue to be used in DNA reaction rate
 "benchmark" calculations for communication propagation sensitivity assessments, particularly for those frequency ranges which depend on D region characteristics for propagation acceptability in terms of variations in phase and amplitude.
- 2. Positive-Negative Ion Reactions in Various Atmospheric Gases-F. Niles and J. Vanderhoff, AASLNM and BRL. Ends 30 September 1977 (Work Unit 02).

Laboratory investigations of positive and negative ion processes of importance to communications through disturbed and recovering ionospheres (e.g., in case of nuclear detonations) over the temperature range in the D region of the mesosphere will be conducted. Photodestruction cross-sections for complex cluster ions at photon energies between 1.8 and 3.5 eV using Krypton or Argon laser lines will be employed in these efforts. In addition, determinations of the photodestruction cross-sections for such species for various discrete, incremental photon energies will also be employed. This work will be carried out in consort with adjunct experiments in progress at SRI International, particularly for negative ions.

3. Electron and Cluster Ion Recombinations and Temperature Coefficients—M. Biondi, University of Pittsburgh. Ends 30 September 1977 (Work Unit 03).

Investigations of certain electron and cluster-ion recombination rate coefficients, and their dependence upon the electron temperature will be completed in FY77. Water cluster-ions such as $[H_3O^+ \cdot (H_2O)_n]$, which influence HF propagation through the D region, will be included. Special

laboratory measurements of the formation, breakup and D-region reactions between complex cluster ions (e.g., $[NO^+ \cdot O_2]$ switching into $[O^+ \cdot H_2O]$ type species) to determine rate constants and binding energies at moderate and high temperatures (500 and 1500 K) will be conducted. The reaction between O^+ and N_2 at elevated temperatures (500 to 1500 K) will also be investigated.

 Three Body Debris Metal Ion Interactions—W. Fite, Extranuclear Corp. Ends 30 September 1977 (Work Unit 04).

Laboratory investigations are to be performed of D-region processes involving several typical kinds of debris metal ion species with atmospheric minor constituents; and 3 body reactions with major species, e.g., $A\ell^+ + O_3 \rightarrow A\ell O^+ + O_2$; $A\ell^+ + O_1 \rightarrow A\ell O^+ + M$ (similarly with Fe⁺ in lieu of $A\ell^+$). Such reactions are important in late time debris effects since they influence the energies, distributions, and total concentrations of free electrons in the ionosphere. These in turn affect the refractive indices of disturbed D-regions and hence, EM wave propagation.

5. Photodissociation and Photodetachment of Negative Ions and Hydrates-C. Lineberger, JILA. Ends 30 September 1977 (Work Unit 05).

Laser and photoelectron spectroscopic laboratory techniques will be employed to measure bond energies and other important parameters (cross sections, etc.) for hydrated negative ions $[NO_2^- \cdot H_2O]$; $[NO_3^- \cdot H_2O]$; $[CO_3^- \cdot H_2O]$; $[CO_4^- \cdot (H_2O)]$ exposed to radiation in the 6000–2800Å range. Results will be validated by a new laboratory technique which will measure the photoelectron spectra for energetic electrons produced.

6. Positive Ion-Negative Ion Recombination Rate Coefficients for the Lower D Region-J. Peterson, SRI International. Ends 30 September 1977 (Work Unit 06).

Existing merging beam laboratory equipment and procedures will be improved, and new techniques developed, if necessary, to obtain rate constants for positive ion-negative ion, complex and water cluster species reactions applicable to D region, late time ionization processes. The classic reaction of NO⁺ with NO₂ will be used to evaluate the improvements achieved. Subsequently, rate constants will be obtained for positive ion-negative ion neutralization reactions involving such positive ions as NO⁺; O₂⁺; and $[H_3O^+ \cdot (H_2O)_n]$, and such negative ions as O⁻, O₂⁻; $[NO_3^- \cdot (H_2O)_n]$; $[CO_3^- \cdot (H_2O)_n]$; and $[CO_4^- \cdot (H_2O)_n]$. These measurements must be made as a function of ion temperature and under such conditions that the reacting ions and products of importance to the propagation of low frequency bands are unambiguously identified. This information will complement related theoretical calculations described under Subtask S99QAXHDO28, Work Unit 46 (for FY77).

7. E and F Region Rate Coefficients for Complex Negative and Excited Positive Ions-J. Paulson and E. Murad, AFGL. Ends 30 September 1977 (Work Unit 07).

Laboratory measurements of photodissociation rates for positive ions such as N_2O^+ ; $[NO^+ \cdot H_2O]$; and $[O_2^+ \cdot H_2O]$ will be investigated. In addition the rate of the reaction of $O^+ + N_2 \rightarrow NO^+ + N$ with vibrational states of the reactants corresponding to thermal temperatures up to 2200 K will be determined. Knowledge of these interactions is essential for a comprehension of E and F region ionospheric processes affecting the propagation of higher communication frequencies through these upper altitude regions, as well as induced IR emissions.

8. Laboratory Measurements of Selected D-Region Reactions-E.E. Ferguson, et al., NOAA. Ends 30 September 1977 (Work Unit 08).

Several specific problems of importance to DNA upon which work will be done are outlined below. In addition, as new problems develop during the coming fiscal year, they will be investigated.

The D-region positive ion reaction chemistry scheme is incomplete. One important gap in understanding the D-region positive ion chemistry concerns the mechanisms involving NO^+ . The successive hydration of NO^+ by three-body association with H_2O is too slow to account for the observed dominance of H_3O^+ and its hydrates. An alternative mechanism would involve the association of NO^+ and $NO^+(H_2O)_n$, n=1,2 with N_2 and CO_2 to form weakly bound association complexes, which in turn react with H_2O to effect a more rapid hydration of NO^+ . This mechanism will be investigated in the variable temperature flowing afterglow at the appropriate D-region temperature.

The measurement of the important D-region negative ion chemistry will be continued. The reaction rate constants of the reactions of O_3^- , O_3^- , O_4^- , and O_4^- with NO, and O_3^- and O_4^- with CO₂ are found to have a strong energy dependence. A similar behavior is expected for the rate constants determined in the temperature variable system.

The flow-drift system will be used to obtain the energy dependence of rate constants for important atmospheric reactions from near room temperature to relative kinetic energies of a few electron volts. The reaction studies at suprathermal energies are important in F-region ion chemistry and under disturbed conditions. Reactions involving atomic oxygen, atomic nitrogen, and vibrationally excited nitrogen will be undertaken in this system.

The mobility of atmospheric ions in various carrier gases will be measured in the flow-drift system. The mobility is important in determining the transport of ions in electric fields. Also, from the zero-field mobility the ion diffusion coefficient in the particular carrier gas may be calculated.

Work will continue on the thermochemistry of atmospheric ions. Knowledge of the heats of formation of these ions is necessary to place precise constraints on the possible reaction paths leading to the production or destruction of these ions. The heat of formation of NO_3 was determined, and that of CO_3 will be studied.

An atmospheric pressure ion source has been constructed and coupled to a mass spectrometer. An investigation of the ions generated in this source can indicate the processes that are important in the ion chemistry of the lower atmosphere and the sensitivity of this chemistry to various trace constituents. The properties of this device will be explored and its application to high-pressure ion chemistry will be examined.

9. Atmospheric Chemical Sensitivity and Modeling Investigations-M. Scheibe, MRC. Ends 30 September 1977 (Work Unit 09).

Mission Research Corporation will:

- Examine certain chemistry code routines to determine the effect of the variation of parameters upon UHF satellite communications systems performance.
- Update the Effective Rate Constants (ERCs) for use in the chemistry computer code models.
- Maintin and update chemistry computer codes and provide "benchmark" calculations to facilitate code improvements.
- Use chemistry computer code for D-region sensitivity studies pertaining to ELF, VLF, and HF communications systems
- Advise and interact, as requested, with special DNA chemistry ad hoc panels and groups.
- 10. Low Energy Cross Sections for Debris Metal Ions-R. Neynaber, D. Vroom, and J.A. Rutherford, IRT, Inc. Ends 30 September 1977 (Work Unit 10).

Data needed for codes to model upper atmospheric disturbances will be obtained. The following tasks will be included:

- The reaction N⁺ + O → N + O⁺ will be investigated using crossed beam techniques. The
 cross section or an upper limit on the cross section will be determined at the lowest possible energy obtainable.
- The reactions

$$Al^{+} + O_{2} = AlO^{+} + O_{2} + Al + O_{2}^{+}$$

and

$$A\ell^{+} + N_{2}$$
 $A\ell^{+} + N_{2}$
 $A\ell^{+} + N_{2}^{+}$
 $A\ell^{+} + N_{2}^{+}$
 $A\ell^{+} + N_{2}^{+}$

will be investigated using crossed beam techniques. The energy range of the measurements will be 1 to 5000 eV.

11. Improved Master/Simple Codes for D, E, F Regions-W. Ali, NRL. Ends 30 September 1977 (Work Unit 11).

The following tasks, under the direction of W. Ali at the Naval Research Laboratory will be performed:

- Maintain and update the NRL "MASTER" and "SIMPLE" computer codes through use of new and/or updated ionospheric reaction rate information.
- Participate in DNA-sponsored "benchmark" activities designed to acquire improved effective rate coefficients for use in ionospheric chemistry/communications codes sensitivity assessment.
- Provide essential reaction rate information to other DNA-sponsored NRL activities engaged in improvement to the present UV fireball and other elements of NRL communications/ optical codes.

B. SUBTASK S99QAXHD028

"Theoretical Investigations of Ionizing Mechanisms in the Upper Atmosphere"

1. Computations of Molecular Structures and Transition Probabilities—H. Michels, UTRC. Ends 30 September 1977 (Work Unit 44).

Computations of atomic structures and transition probabilities will be made (e.g., NO⁺ ion), which in turn permit calculations of detailed electronic energy levels, potential energy curves, and radiative lifetimes. Subsequently, calculations for the reaction N⁺ + O \rightarrow N + O⁺ at low energies (0-5 eV) will be conducted to supplement experimental work by IRT, Inc., as described under subtask S99QAXHD411. Such calculations will specifically involve examination of the following excited species: N⁺(3P) + O(3P) \rightarrow N(4S) + O⁺(4S) and N⁺(1D) + O(3P) \rightarrow N(2D) + O⁺(4S). The reaction rate for NO⁺ + e \rightarrow N(2D) + O will also be recalculated by an improved procedure to serve as a check against new experimental data.

 Theoretical Aspect of Ion-Ion Laboratory Measurements-F. Smith, SRI International. Ends 30 September 1977 (Work Unit 45).

Calculations of parameters affecting the role of clustered ions in neutralization reactions will be performed. Semi-classical techniques will be modified in rate calculations to account for effects resulting from the increase of internal degrees of freedom in such species. Techniques will be developed to

calculate recombination rate coefficients for ionic species such as $[H^+ \cdot (H_2O)_n]$; $[NO^+ \cdot (H_2O)_n]$; $[NO^- \cdot (H_2O)_n]$; $[NO_3^- \cdot (H_2O)_n]$.

 Theoretical Investigations of Disturbed Ionospheres—W. Swider, AFGL. Ends 30 September 1977 (Work Unit 46).

Data from SPE 1972 will be assessed against SPE (PCA) 69 results to try to account for the very high electron loss ratio measured at 80 km in the D region. Similarly, the SPE 72 data will be compared with related parameters and models deduced from SPE (PCA) 69 to note discrepancies and agreements. Subsequent efforts will concern efforts to elucidate the role of NO⁺ ions in auroras, in terms of total positive ion concentration in the E region, and an investigation and documentation of information relevant to satellite communication problems currently available on data tapes from the NASA-sponsored "Atmospheric Explorer" satellite which is available and believed to contain information concerning diurnal chemistry and physics data of interest to DNA.

4. Atmospheric Phenomena Associated with Nuclear Bursts-E. Bauer, IDA. Ends 30 September 1977 (Work Unit 47).

High-altitude nuclear explosions produce large numbers of ions and excited atoms and molecules, which can pose a severe threat to the operation of survivable communications and other high priority DoD defensive systems. The Defense Nuclear Agency is currently engaged in investigations covering the significant parameters and phenomena relevant to this situation through a series of coordinated theoretical, laboratory, and field measurement programs. IDA has reviewed pertinent geophysical phenomena which can be utilized to effectively simulate specific aspects of high altitude nuclear explosions on the performance of critical DOD communication systems.

 Reaction Rate Sensitivity Assessments—M. Bortner, T. Baurer, GEMSD. Ends 30 September 1977 (Work Unit 48).

In cooperation with the DNA/COR, the necessary activities of DNA Special Ad Hoc Panel Number 2 will be conducted. This involves the identification and assessment of ionospheric chemical and physical parameters which influence the output of computer codes. The Panel members working toward these objectives will be monitored and assisted.

Progress made in related technical areas will be assessed and utilized to ensure completion of existing and/or planned DoD communications system mission objectives.

Also final revisions to the DNA Reaction Rate Handbook, Second Edition (DNA 1948H), will be edited and additional Handbook revisions provided to maintain its capabilities in the various fields covered, with particular concern for its relevance to evolving DNA/DoD/communications systems requirements.

C. SUBTASK S99QAXHI002

"Atomic and Molecular Physics of IR Emissions"

1. Chemistry and Spectroscopy of Optical Emitters – D. Snider and F. Niles, AASLNM. Ends 30 September 1977 (Work Unit 41).

The acquisition, evaluation, and assessment of lower D region (45-65 km) electrical and chemical parameters, including ion conductivities, ion-mobilities, photo-induced ion formation, and reactions during daytime high-altitude disturbed environments are planned. Measurements will be made with appropriate ground-based, rocket-borne, and balloon-borne instrumentation in Alaska in mid Calendar Year 1977. Extension of results to various high altitude nuclear detonation disturbed ionospheres will also be made where data permit.

Lower D region atmospheric data will be obtained from the balloon-borne platforms with suitable optical instrumentation in order to delineate more specifically the nature and possible sources of infrared emissions which have been observed in past Alaskan field measurements. Correlation of such data with that described in the above paragraph will be made where possible in order to elucidate how daytime disturbed ionospheric parameters compare with the more intensive and usual types of night-time disturbances.

2. UV and VUV Photoabsorption and Photoionization Investigations-R. Huffman, AFGL. Ends 30 September 1977 (Work Unit 42).

The compilation of ionospheric photodissociation rate data for the DNA Reaction Rate Handbook will be completed. A brief survey of ultraviolet absorption bands from vibrationally excited ground state molecular nitrogen will be done to determine which bands absorb UV fireball emissions (work will be done in coordination with NRL and RDA, Inc.).

Will also evaluate and select specific UV imaging detectors which can be used to characterize and elucidate ionospheric irregularities relevant to effects upon satellite communication systems.

A complete assessment of the effects of UV fireball emission lines upon the extent of photoionization in the F-region of the ionosphere will be performed. Finally, designated UV and VUV, plus related optical sensor/imaging instrumentation will be identified which can be effectively employed in possible future space shuttle flights of DoD/DNA interest.

3. Assessment of Oxygen and Ozone Data from Disturbed Environments-L. Weeks, and K. Champion, Boston College/AFGL. Ends 30 September 1977 (Work Unit 43).

The reduction and assessment of data obtained in-situ in the lower D region during a magneto-spheric-substorm at high latitudes (Alaska) in FY 76 will be completed. Data were obtained for variations produced in the concentrations of O₂, O₃, and total density. Results will be used to supplement similar data obtained from the SPE (PCA) 69 and 72 events, in order to improve the daytime model for the D region under highly disturbed conditions. These results will also be extended to possible nuclear situations.

4. Reactions of Excited Atmospheric Gases-F. Kaufman, University of Pittsburgh. Ends 30 September 1977 (Work Unit 44).

In FY 77 laboratory investigations applicable to disturbed ionospheres will be conducted to obtain the following information: (1) Vibrational excitation and relaxation of NO, CO, CO₂ molecules as determined from fast flow, IR chemiluminescence. Vibrational relaxation rate constants for atmospheric quenching molecules (N_2 , O_2 , CO_2) will be measured up to high v states of the excited molecules. (2) Electronic and vibrational excitation in air triatomics (such as NO_2) will be measured by tunable dye laser absorption. From the spectrally resolved time decay of the fluorescence intensity in the presence and absence of quenching molecules, detailed mechanisms of energy disposal in the 2 to 3 eV excitation energy range will be mapped out. (3) Reaction and quenching processes of $N(^2D)$ and $N(^2P)$ species with $O(^1D)$, O_2 , and CO_2 will be investigated by resonance fluorescence methods with emphasis on elucidation of product channels. (4) The role of the so called "A" state of nitrogen in excited state reactions involving N atoms will also be investigated.

5. Vibrational Energy Transfer Investigations and Spectral Data for UO⁺-F. Bien and M. Camac, Aerodyne Research Laboratories. Ends 30 September 1977 (Work Unit 45).

A comparison of IR data from previous U.S. atmospheric high altitude nuclear tests will be made with the laboratory measurements which were conducted in FY 7T for the f-number of excited state NO⁺ emissions at 4.3 microns. Additionally, investigations concerning the vibrational lifetime of

excited NO⁺ (for v=1, 2, 3) and the transfer of that vibrational energy to N_2 and O_2 will be completed. Laboratory measurements to determine IR spectral data for UO and UO⁺ species (not obtainable for UO⁺ with the Argonne Laboratory matrix isolation technique) will be made to complete the spectral atlas for uranium oxide species of nuclear debris interest.

 Optical IR Sensitivity Assessments—D. Archer and D. Sappenfield, Mission Research Corporation. Ends 30 September 1977 (Work Unit 46).

The work to be performed is directed toward identifying key parameters that should be measured, and/or problems that need to be solved, to improve the accuracy with which IR radiation in a nuclear environment can be predicted, especially in chemiluminescent and vibraluminescent bands between 2 and 5 μ m that impact on space detection and assessment systems. This work shall include, but not necessarily be limited to, the following items:

- From discussions with workers in the field, and from literature surveys, determine the current state of knowledge on factors related to chemiluminescent and vibraluminescent emission in bands near 2.7 and 4.3 μm. The factors would include values and associated uncertainties of reaction rate constants, photons per chemiluminescent reaction, band intensities, mixed mode excitation of polyatomic species, and quenching rates for the relevant vibrational states. Species considered would include NO, OH, HO₂, HNO₃, HNO₂, H₂O₂, CO₂, and NO⁺.
- Where necessary, use available field measurement data along with theoretical analyses, to help resolve uncertainties related to radiating species, excitation mechanisms, etc.
- Attempt to isolate the dominant radiators and excitation mechanisms in the 2.7 and 4.3 µm bands for regions external to nuclear fireballs in the lower D to upper E layers.
- Evaluate the need for, and feasibility of, additional work that would lead to the achievement of a specified prediction accuracy in the 2.7 and 4.3 μm bands.
- Participate, as requested by the Contracting Officer's Representative (COR), within the limits of time and funding, in discussions with DNA and DNA contractors on questions relating to the present program.

D. SUBTASK S99QAXHI004

"IR Phenomenology and Optical Code Data Base"

 Investigations of Specie Structure in Simulated CO₂ Emissions-J. Kumer and T. James, LMSC-PA, Ends 30 September 1977 (Work Unit 27).

Laboratory and data evaluations will be performed to gain a capability for the prediction of spectral, temporal, and spatial structure in the enhanced atmospheric CO₂ infrared radiance expected to occur as the result of energy deposited by non-nuclear mechanisms which simulate the nuclear case.

- Task 1, field data evaluation, is to:
 - a. Develop the numerical capability to calculate auroral CO₂ 4.3 μm zenith spectral radiance.
 - b. Validate this capability via comparison with spectral radiance data which have been obtained via instrumentation in the DNA/AFGL auroral measurements program.
 - c. Elucidate the 4.3 µm auroral zenith spectral radiance results.

• Task 2, laboratory effort, is to:

- a. Measure the 4.3 μm radiance of CO₂ produced in the laboratory by irradiation with 2.7 μm radiation under a range of pressures of CO₂ with varying amounts of rare gases, Ne, Ar, He. Conditions will be chosen so as to produce data over a range of line shapes which vary from a nearly pure Doppler line shape to a pressure-broadened Voight profile with a width several times the Doppler width.
- b. Measure the effects of a small admixture of N_2 on the 4.3 μ m radiance and evaluate these results in terms of the vibrational energy transfer between CO₂ and N_2 .
- c. Evaluate the data obtained in the above two tasks to determine the vibrational transitions involved in producing the observed radiance, and to determine the effects of varying line shapes on the effectiveness of 2.7 μ m radiation in producing 4.3 μ m radiance.
- d. Relate the results obtained with varying line shapes and pressure to the altitude dependence of the atmospheric $4.3 \mu m$ radiance.
- e. Measure the 4.3 μ m radiance at reduced temperatures and utilize these results to determine the contribution of hot bands to the observed signals.
- f. Based on the results obtained in the above steps, investigate the usefulness and desirability of utilizing a laser at 10.6 μ m and 2.7 μ m to produce 4.3 μ m emission. Make recommendation as to whether or not such additional efforts would contribute significantly to an understanding of nuclear induced CO₂ radiance.
- g. The results of the field data evaluation effort and the laboratory effort will be combined into a composite final report.

E. SUBTASK 125BAXHX632

"IR Phenomenology and Optical Code Data Base"

 LABCEDE-Investigations of Irradiated N₂, O₂ and Other Gas Mixtures-R. Murphy, AFGL. Ends 30 September 1977 (Work Unit 07).

The large amount of experimental data obtained in recent months with the operational LABCEDE facility will be reduced and assessed. Work emphasis should be placed upon cleanup of mechanisms concerned with prominent SWIR and LWIR emissions resulting from the irradiation of N_2 , O_2 , and other gaseous mixtures when irradiated with energetic (kev) electron beams.

COCHISE-Investigations of Ozone and Other Reactions of E Region Importance

–J. Kennealy and F. Del Greco, AFGL. Ends 30 September 1977 (Work Unit 08).

The backlog of experimental data obtained in recent months with the COCHISE facility will be reduced and assessed. Work emphasis will be placed upon classification of the mechanisms (e.g., $\bar{v} - \bar{v}$ transitions, quenching, etc.) which contribute to the infrared emissions of a gaseous chemical species which are prominent in the SWIR and LWIR areas of the spectrum in disturbed environments.

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